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Precision and Accuracy of a Beta Gauge for Aerosol Mass Determinations

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Results of an experimental determination of the precision and accuracy of a beta ray attenuation method for measurement of aerosol mass are presented. The instrumental precision for a short-term experiment was 25 µg for a 6.5 cm^2 deposit collected on approximately 1 mg/cm^2 Teflon filters; for a longer term experiment the precision was 27 µg. The precision of the gravimetric determinations of aerosol deposits was 22 µg for Teflon filters weighted to µg. Filter re-orientation and air density changes that were able to adversely affect the beta ray attenuation results are discussed. Beta ray attenuation results are in good agreement with gravimetric measurements on the same filter-collected aerosols. Using dichotomous samplers in Durham, NC, aerosols were collected on Teflon filters in two size ranges. A regression line was calculated implicitly assuming errors in both measurements of mass. The 90% confidence intervals lay within 21 µg of the regression line for mean fine fraction aerosol mass loadings of 532 µg and within 15 µg of the regression line for mean coarse fraction aerosol mass loadings of 392 µg.

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Precision and Accuracy of a β Gauge for Aerosol Mass Determinations

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■ Results of an experimental determination of the precision and the accuracy of a β-ray attenuation method for measurement of aerosol mass are presented. The instrumental precision for a short-term experiment was 25 μg for a 6.5-cm^2 deposit collected on approximately 1 mg/cm^2 Teflon filters; for a longer-term experiment the precision was 27 μg. The precision of the gravimetric determinations of aerosol deposits was 22 μg for Teflon filters weighed to 1 μg. Filter reorientation and air density changes that were able adversely to affect the β-ray attenuation results are discussed. β-ray attenuation results are in good agreement with gravimetric measurements on the same filter-collected aerosols. Using dichotomous samplers in Durham, NC, we collected 136 aerosol samples on Teflon filters in two size ranges. A regression line was calculated implicitly assuming errors in both measurements of mass. The 90% confidence intervals lay within 21 μg of the regression line for mean fine fraction aerosol mass loadings of 536 μg and within 19 μg of the regression line for mean coarse fraction aerosol mass loadings of 349 μg. Any bias between gravimetric and β-gauge mass measurements was found to be less than 5%.

Introduction

One of the most widely reported parameters of atmospheric aerosols has been the mass concentration deduced from particulate matter collected on filters. In such measurements mass is usually determined by weighing. Gravimetric procedures, however, are inconvenient for analyzing large numbers of samples, since such procedures require a significant amount of sample handling and are difficult to automate. Jaklevic et al. have recently described the design of a β gauge for determining the mass of aerosols collected in dichotomous samplers on thin, Teflon membrane filters (*1*). The β gauge is a device consisting of a β-ray source, a β-ray detector, and counting electronics and is used to deduce the mass of an object placed between the source and the detector. Mass measurements by β gauge have been reported in several previous studies on atmospheric aerosols (*2–5*) and this paper provides experimental justification for the assumption of equivalence of gravimetric and β-gauge measurements of mass.

In this work we report experiments designed not only to test the equivalency of a β-gauge method to the standard gravimetric determination of aerosol mass but also to determine the operational characteristics of a β gauge. The filters used were Teflon membranes, and the aerosols were collected in two size ranges by using dichotomous samplers. Results are presented for experimental determinations of precision and accuracy of gravimetric and β-gauge measurements. Studies of the effect of filter orientation and laboratory atmospheric conditions on β-gauge response are described. These experiments show that, if filter orientation is changed between the measurement of the blank filter and the loaded filter, the precision of the β-gauge mass determination suffers significantly. The effects of laboratory atmospheric conditions are also shown to be important.

Experimental Procedures

Filters. The filters were unbacked, Teflon membranes having 1-μm pore size and were used in two different types of mounts. In one, the filters were heat sealed onto supporting annular polyolefin rings; in the other they were heat sealed onto (5 × 5) cm^2 plastic frames. Both filter types are standard for use in Beckman automated dichotomous samplers and were obtained from Ghia Corp., Pleasanton, CA, and from Beckman Instruments, Fullerton, CA. The area of the aerosol deposits in these experiments was 6.5 cm^2. The filter material had an areal density of about 1 mg/cm^2.

Gravimetry. Mass measurements were made by using a Mettler ME22 balance with a BE22 control unit. Absolute accuracy was determined by using a new set of class S weights (*6*). The polyolefin-ringed Teflon filters used in this experiment had masses of approximately 100 mg; the tolerance for a 100-mg class S weight was 15 μg. Calibration of the balance over the range 0–100 mg showed that its *absolute* error was no more than 21 μg. A ^{210}Po source was used to neutralize static electrical charge on the filters during gravimetric measurements.

β-Gauge Measurements. The β-gauge system used in

the measurements reported here was designed and fabricated by Lawrence Berkeley Laboratory and is described by Jaklevic et al. (*1*). The response of a β gauge is described by the equation

$$I = I_0 e^{-\mu x} \quad (1)$$

where x is the areal density (mass per unit area) of the sample being measured, μ is the mass absorption coefficient for β radiation in the sample, I is the measured count rate when the sample is placed between the source and the detector, and I_0 is the count rate when no sample is present. When the coefficients I_0 and μ are known, one can determine x for an unknown sample by measuring I and using eq 1.

The β-ray source was ^{147}Pm ($t_{1/2}$ = 2.623 yr; $E_{\beta max}$ = 224 keV) and had an activity of 1 mCi. The diameter of the active area was 2.2 cm. The detector was a silicon surface barrier detector operated at room temperature. The active area of the detector was 3.0 cm^2 with a 300-μm sensitive depth. The source-to-detector distance was 0.7 cm. We performed a Monte Carlo calculation to estimate that the average path length of β-rays from source to detector was 1.22 cm. The sample holder contained four cartridges. The top and bottom ones had six positions each and contained standards and blank samples. The standards consisted of an empty frame and five polycarbonate films which have known areal densities. The calibration standards spanned the areal density region of 684–1866 $\mu g/cm^2$. The blank samples were five Teflon membrane filters (1-μm pore size) of the type used in most of our ambient aerosol sampling studies and one frame without a filter. Each of the middle two cartridges contained 36 unknown samples.

Counting times of 90 s were used for all measurements reported in this work. The 84 individual 90-s analyses automatically are done over 2.1 h. The count rate ranged from 3×10^4 to 10^5 Hz. For each set of mass measurements, a calibration was derived from the observed counts for the five polycarbonate standards. The calibration parameters in I_0 and μ were determined from a least-squares fit using eq 1 in linear form:

$$\ln I = \ln I_0 - \mu x \quad (2)$$

For each unknown sample the aerosol deposit mass x was determined from eq 2 using paired measurements of I that were made both before and after the aerosol was deposited.

Validation of the analysis was based on operational data. The count rate through an empty frame was obtained at the beginning and the end of the analysis period and tested for agreement to within Poisson counting statistics (i.e., ± square root of the counts). Areal densities of the five blank Teflon filters were compared with their individual cumulative mean values. The cumulative standard deviation was used to validate the analysis in the following manner: if more than one of the blank sample areal densities differed from the cumulative mean by twice the cumulative standard deviation, the entire analysis was invalidated and repeated. During routine operation of the β gauge from November 1978 to November 1980 fewer than 2% of the analyses had to be invalidated. The causes were identified as operator error (e.g., entering incorrectly the areal densities of the calibration standards) and atmospheric density changes (discussed in detail in a later section).

Results and Discussion

Precision of Gravimetric Measurements. The precision of the gravimetric mass determinations was evaluated by analysis of two data sets. The first set was composed of 150 replicate weighings of a polyolefin-ringed Teflon filter over a 2-yr period. This filter was stored in a petri dish between weighings. The mean of the mass determination was 111.3617 mg with a standard deviation of 0.0083 mg. From these results and assuming that both initial and final weights were of the same precision, we estimate 11.7 μg for the precision of gravimetric determination of filter-collected aerosol mass.

Table I. Precision of Gravimetric and β-Gauge Measurements Using Teflon Membrane Filters

expt	no. of filters	mean difference, μg	SD of difference, μg
gravimetry	50	−2	22
β gauge short-term	72	−5[a]	25[a]

β-gauge long-term filter no.	no. of measurements	mean areal density, $\mu g/cm^2$	SD,[a] μg
1	86	1058.3	32
2	86	1040.2	30
3	86	1137.5	26
4	86	1301.5	23
5	86	659.8	25

[a] Calculated for a 6.5-cm^2 deposit area.

The second set of measurements was designed to simulate handling of Teflon filters in ambient aerosol sampling experiments. Blank Teflon membrane filters with supporting rings were weighed and then mounted in frames for use in an aerosol sampler. The filters were then run through an analysis in the β gauge, demounted, and weighed again. The results are presented in Table I. The mean difference in the pairs of initial and final weights was −2 μg, and the standard deviation of the differences was 22 μg.

The precision estimate based on the second set of data is believed to be the best available estimator of the precision of gravimetric determination of the aerosol mass collected on Teflon filters. Factors which may have contributed to the worse precision in the second estimate included sample handling by both the balance operator and the automated equipment, poor reproducibility of static electric charge neutralization, and changes in the ambient weighing conditions.

Precision of β-Gauge Measurement. Short-term and long-term experiments were carried out to test β-gauge precision. In the short-term experiment, 72 Teflon filters bonded in frames were measured twice on the same day. The mean and the standard deviation of the differences in the paired mass determinations are given in Table I. In the long-term experiment, five filters were each measured 86 times over a period of 1 yr. The reproducibility of determinations for each filter is indicated by the standard deviations shown in Table I. Short-term results show that the precision of the β gauge was 25 μg (or 3.8 $\mu g/cm^2$ for 6.5-cm^2 area), which is comparable to our gravimetric determinations. The β-gauge precision over a 1-yr period was 27 μg.

Orientation of the Filter in the β-Gauge. Visual inspection of the Teflon membrane filters revealed a pattern, or grain, running predominantly in one direction. Microscopic inspection revealed individual fibers running predominantly in another direction. The two patterns were often perpendicular to one another. The orientation of a filter in the β gauge would affect the β-ray attenuation if the filter and/or the β source and/or the β detector response were not uniform. To determine the effect of

Table II. Effect of Filter Orientation on Reproducibility[a]

	SD, %	50% confidence interval half-width %	50% confidence interval half-width μg/cm²	90% confidence interval half-width %	90% confidence interval half-width μg/cm²
filter at 0°,	±0.37	±0.26	±2.6	±0.67	±6.4
filter at 90°, 180°, 270°	1.8	±1.2	12	±3.1	±31

[a] The values in μg/cm² are calculated for filters having 1 mg/cm² mass per unit area.

filter orientation, we measured filter masses at orientations of 0°, 90°, 180°, and 270°. These orientations were achieved simply by rotating the filter between measurements; the filter plane was, of course, preserved perpendicular to the β-ray path.

Six blank polyolefin-ringed Teflon membrane filters were each measured in the β gauge 4 times in succession at an orientation of 0°. The standard deviation for these six groups of measurements ranged from 0.17% to 0.62% of the mean values. Normalizing each mean to 1.00 allowed computation of the standard deviation of the entire set of measurements: it was 0.37%. To determine the appropriate confidence interval, we used the t coefficient for 18 degrees of freedom. (The number of degrees of freedom is given by the number of measurements minus the number of mean values determined (24 − 6 = 18).) Ranges corresponding to 50% and 90% confidence intervals are given in Table II. These results indicate that, for two successive measurements of the same filter without changing orientation, the chances were 50% that the agreement would be better than 2.6 μg/cm² and 90% that the agreement would be better than 6.4 μg/cm².

For the set of six filters, the masses determined at 90°, 180°, and 270° ranged from 96% to 102% of the mean mass determined at 0°, and the standard deviation was 1.8%. When one compares the confidence intervals for repeated measurements at the same orientation (0°) with those at different orientations, it is clear that the precision of mass determination using the β gauge can be considerably worse if the orientation of the filter in the β gauge is changed between the initial and final measurement. The results showed no consistent pattern of change in β-ray attenuation when the filter was oriented with respect to either visual or microscopic filter grain.

Effects of Changes in Air Density. The average path length of a β particle in the gap between source and detector was 1.22 cm. The corresponding areal density of the air in the gap was 1.6 mg/cm² at a temperature of 20 °C and a pressure of 760 mmHg. Although the areal density of air in the gap was fairly large, its effect upon our measurements was usually eliminated by the calibration procedure. However, in one instance an operator error caused a 12-h interval to occur between calibration and analysis of samples. During that time, a frontal passage caused the pressure to decrease by almost 6 mmHg. For a 1.22-cm air path the corresponding decrease in areal density was 11.5 μg/cm². For the five quality control filters, the observed average areal density determined by the β gauge was too low by 9.1 μg/cm².

The previous example demonstrates the importance of the quality assurance procedure. If the calibration is carried out immediately before mass determination, then short-term local room-temperature variations are more important than changing weather. For example, if the room temperature varies by ±1 °C, then air density will vary by ±4.1 μg/cm³; this variation is equivalent to ±5.8 μg/cm² for a 1.22-cm air gap.

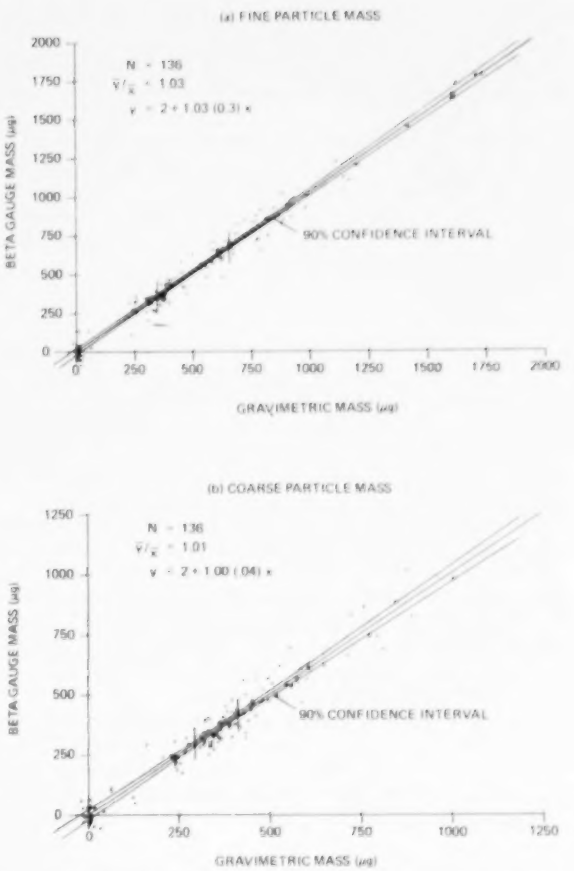

Figure 1. Comparison of β-gauge and gravimetric mass determinations for ambient aerosols collected in two size fractions: (a) results for particles with aerodynamic diameter less than 2.5 μm; (b) results for particles with aerodynamic diameter in the range 2.5–ca. 15 μm. The fitting procedure and confidence interval estimates are discussed in the text.

Comparison of Aerosol Mass Determinations by β Gauge and Microbalance. We minimized handling errors and eliminated orientation effects by carrying out the experiment in the following sequence: (1) measure gravimetric tare mass, (2) measure β-gauge initial areal density, (3) collect aerosol, (4) measure β-gauge final areal density, (5) measure gravimetric final mass. The aerosols were collected on Teflon filters bonded to polyolefin rings using three automated dichotomous samplers. The study site was on the roof of a one-story building in downtown Durham, NC. The samples were collected during 1980–1981 using 24-h collection periods.

Results of aerosol mass determinations on 136 fine and 136 coarse fraction samples are presented in Figure 1. The linear regression lines shown in Figure 1 were computed by a technique which is appropriate when experimental error exists in both the x and y coordinate data (7). The 90% confidence intervals of the slopes are shown in parentheses. Figure 1 also shows boundaries of the 90% confidence intervals for all values of ordinate and abscissa. The indicated 90% confidence interval is actually a conservative estimate. The 90% confidence intervals lie within 21 μg of the regression line for mean fine fraction aerosol mass loadings of 536 μg (82 μg/cm²) and within 19 μg of the regression line for mean coarse fraction aerosol

mass loadings of 349 μg (54 μg/cm²). As shown in Figure 1, the ratios of means are 1.03 and 1.01 for fine and coarse fractions, respectively, indicating that any bias between gravimetric and β-gauge mass measurements is less than 5%.

Based on X-ray fluorescence elemental analysis of these filters, the aerosol composition at the study site was similar to that observed in other urban areas (2–5, 8); consequently, our observed measures of agreement between gravimetric and β-gauge masses should be representative of urban samples, generally.

Conclusions

The data presented here permit several conclusions about the use of β attenuation to determine the mass of particulates collected on filters. (1) Ambient aerosol mass determinations by β attenuation are in good agreement with gravimetric determinations on the same Teflon membrane filters. Any bias between gravimetric and β-gauge mass measurements was found to be less than 5%. (2) The precision and the long-term stability of β-gauge mass measurements can be comparable to that achieved with a microbalance. (3) To obtain precise aerosol mass measurements using a β gauge, the filter orientation must be the same for initial and final measurements. (4) To obtain precise results using the β gauge, the air density cannot change appreciably between the time that the standards and samples are analyzed.

The problems in maintaining constant filter orientation and air density can be overcome by careful handling and operation in a temperature-stabilized environment. The gains achieved by using the fully automated β gauge are reductions in sample handling and operator time, which make β-ray attenuation an attractive alternative to gravimetric assay of large numbers of filters.

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